

Fermi and Intramode Relaxation Phenomena in CO₂ Lasers

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Abstract—Using a 1 ns pulse from a short-pulse CO₂ laser system, the evolution of the gain in a TEA system was studied during and after amplification. This resulted in a very direct observation of a few relaxation processes. We estimated the effective intramode relaxation rate constant to be larger than $6 \times 10^6 \text{ torr}^{-1}/\text{s}$. The Fermi relaxation time constant was found to be $30 \pm 7 \text{ ns}$ at 760 torr. We conclude that for nanosecond pulse amplification, intramode relaxation cannot be neglected.

I. INTRODUCTION

IN principle, nanosecond pulses are less efficiently amplified in a CO₂ laser amplifier than pulses that are much longer. This is caused mainly by the limiting rotational relaxation time constant of about $0.15 \text{ ns} \cdot \text{atm}$ [1]. To overcome this problem, several techniques can be employed. The first is to increase the pressure of the laser gas because the relaxation time is inversely proportional to the pressure. However, gas discharges at higher pressures are more difficult to control. The second, widely investigated, technique is to extract energy from the amplifier by stimulated emission on many rotational lines or even on two bands [2]–[4]. The results obtained by this technique depend on various conditions, such as the ratio of the pulse duration to the rotational relaxation time and the intensity of the incident pulse. Obviously, for weak pulses not affecting the gain, multiline energy extraction is useless. On the other hand, even multiline extraction in a vibrational band can use only the available energy stored in that band. The effect of these techniques is to extract the available energy from all rotational levels. For a 1 ns pulse, the effect of multiline extraction is an increase of about a factor of 2 [4]. We notice, however, that the effect is less if the beam profile is not uniform, for example, Gaussian. In that case, the low-intensity regions experience little advantage from the multiline extraction technique. A second limitation on efficient amplification of nanosecond pulses may be intramode relaxation. During amplification, the interaction of the radiation field is, for instance, between the (00°1) and (10°0) vibrational levels of the ν_3 - and ν_1 -vibrational modes, respectively. At first, only these levels are changed, and the population densities of the higher levels of the ν_1 - and ν_3 -vibrational modes are not affected. Since the population densities of

the lasing levels are changed, the original thermal distribution of each vibration mode is disturbed. These disturbances cause intramode relaxations in order to restore the thermal distributions. In the case where the duration of the intramode relaxation process is larger than or comparable to the pulse duration, we are dealing with reduced energy extraction from an amplifier. Next to the intramode relaxation there is also an energy exchange between the ν_1 - and ν_2 -vibrations, i.e., relaxation by Fermi resonance. The relaxation processes in CO₂ or in a CO₂ laser gas mixture have been investigated by various workers (e.g., [5]–[9], [13]). Yet the information is still incomplete. In particular, the rates of the intramode relaxation processes are not accurately known. In the present work, we investigate the short pulse amplification during and after amplification. The observations are analyzed with a theoretical model, and the results contribute to an understanding of Fermi and intramode relaxation phenomena.

II. DIRECT MEASUREMENT OF GAIN RELAXATION

A. Experimental Configuration

The experimental setup that is used for the determination of the time evolution of the gain is shown in Fig. 1. When the nanosecond pulse laser system is turned off, the setup is a conventional gain measurement system. The laser system is synchronized with the rotating chopper wheel. The wheel has only two small apertures, resulting in an optical pulse of 1 ms at the detector every 16 ms. In this way, thermal heating of the detector by the optical radiation is greatly reduced. The detector is a room-temperature, HgCdTe detector, type R006-2, manufactured by Radece. The sensitivity is 40 mV/W, and it has a specified rise time of 0.4 ns. Because of the fact that this detector is very fast, we are able to investigate the recovery of the gain when it is perturbed by the passage of a high-intensity nanosecond pulse. All experiments are performed at atmospheric pressure (760 torr).

As can be seen from Fig. 1, the CW probe laser passes the TEA amplifier at a small angle with respect to the high-intensity pulse. This slight difference in beam directions has only a small diminishing effect on what can be expected from a collinear experiment. The beam radii of the pulsed and probe beam ($1/e^2$ intensity points) are $r_0 = 3.0 \text{ mm}$ and $r_0 = 2 \text{ mm}$, respectively. The angle between the beams is 1.1 deg. The direction of polarization of the probe beam is horizontal and perpendicular to that

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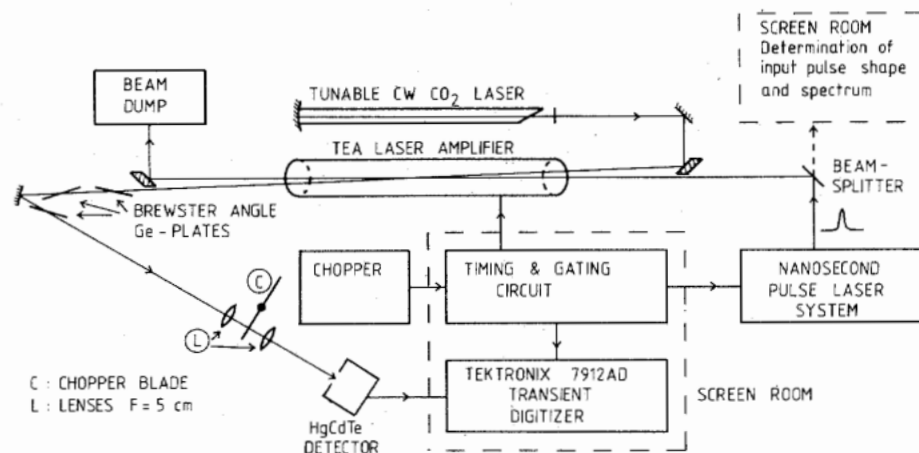


Fig. 1. Experimental configuration for determination of the time evolution of the gain.

of the pulsed beam. One might suggest using two collinear beams and separating them at the exit of the amplifier using Ge Brewster plates. This is, however, not attractive. The expected effect of the high-intensity pulse on the gain signal is on the order of 10 mV. The 1 ns pulse has a power of about 100 MW. Therefore, to obtain a clean signal, the power of the residual pulse reaching the detector must be less than 0.02 W, or 2.10^{-10} times the original power. Even in our setup, we used three plates to attenuate the energy of the pulse reaching the detector at the desired level. In spite of this small deviation of the two beam directions, we may consider that the probe laser radiation travels through the amplifier in practically the same direction as the disturbing pulse. This allows us to follow relaxation processes that occur on a nanosecond time scale. This would not have been the case if the beams had been in opposite directions. The $2 \times 2 \times 40$ cm TEA laser amplifier of the type described by Ernst [10] was operated using a CO₂:N₂:He = 1:1:3 mixture. The energy input was 225 J/L.

B. Experimental Results

The experiments can be divided into a few categories:

- 1) determination of the gain evolution without a disturbance,
- 2) determination of the gain evolution with a pulse of 10 ns duration passing the amplifier at peak gain, and
- 3) As in 2), but with a pulse of 1.0–1.5 ns.

1) *Determination of the Gain Evolution Without a Disturbance:* For this case, the nanosecond pulse laser system is switched off. By properly timing the setup, it is possible to obtain a signal as shown in Fig. 2. From this signal the gain of the laser amplifier is determined to be 3.1 ± 0.1 percent/cm.

2) *Determination of the Gain Evolution with a Pulse of 10 ns Duration Passing the Amplifier at Peak Gain:* In order to investigate the recovery of the gain after a disturbance, we injected a high-intensity pulse into the amplifier at the moment of maximum gain. The results showed at least two relaxation effects: one with a time constant of about 10–40 ns and one with a time constant of about 350 ns. In order to determine the latter, the size

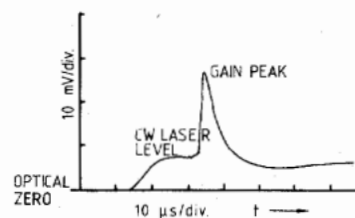


Fig. 2. Gain signal of the TEA CO₂ laser amplifier, showing optical zero, CW optical level, and gain peak. Laser mixture is CO₂:N₂:He = 1:1:3. Electrical input power is 225 J/L.

of the disturbance was made as large as possible. We therefore injected a pulse of 10 ns duration and an energy of 150–200 mJ. A typical result is shown in Fig. 3. In this case, the probe laser as well as the pulsed laser system operate on the $P(20)$ transition of the $10.4 \mu\text{m}$ branch. From five of these measurements, we determined the slower time constant for relaxation of the gain to be 340 ± 40 ns. This relatively slow relaxation process corresponds to the transfer of energy from N₂ to the asymmetric stretch mode of CO₂. The small amount of ringing that is observed in Fig. 3(c) is due to electromagnetic interference with the noise produced by the pulsed discharges. Fig. 3(b) and (c) shows that there is also a relaxation process on a shorter time scale.

Typical examples of traces corresponding to this process are shown in more detail in Fig. 4. In these two cases, probe and pulse lasers were operating on the $P(20)$ transition in the $10.4 \mu\text{m}$ branch. The pulse passes near the point of maximum gain. We averaged over many measurements to find the short relaxation time constants. The results are shown in Table I. We notice that the recovery of the gain on this time scale is only 30–40 percent of the size of the disturbance. Looking in the literature [5] and keeping in mind the order of magnitude of this relaxation time, it is most likely that we are dealing with the Fermi relaxation between the symmetric stretch and the bending modes.

3) *Gain Evolution with a Pulse of 1.0–1.5 ns Passing the Amplifier at Maximum Gain:* When the pulse passing the amplifier has a duration of only 1 ns, it is possible to

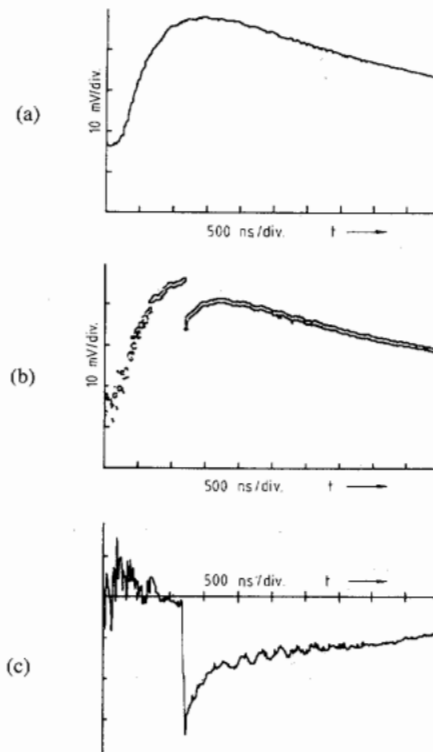


Fig. 3. Gain relaxation. (a) Unperturbed gain signal. (b) Gain signal with a 10 ns, 180 mJ pulse passing the amplifier. (c) Difference between the signals in (b) and (a).

study the relaxation process in more detail. This is shown in Figs. 5 and 6. In Fig. 5, three pulses pass the amplifier at a time interval of 12.5 ns. To be sure that we obtained all information from these experiments, the phenomena were also studied on a 5 ns/div time scale. This shows that even an effect due to rotational relaxation can be observed. In Fig. 6(a), the gain is probed at the same transition as that of the pulses. A sharp decrease in the gain is observed as the pulse passes. As soon as the pulse intensity decreases, there is a slight but fast (within ~ 2 ns) rise. On the other hand, if the gain is probed on another transition [see Fig. 6(b)], the decrease is slower and continues 1–2 ns after the pulse has passed. This is in agreement with the theory of rotational relaxation.

III. THEORETICAL MODEL

In order to analyze the results, we compare them to the results of a numerical model, which will now be described. The entire model, which will be called the “extended” model, consists of two parts:

- 1) a five-temperature model of CO_2 laser dynamics, and
- 2) a reservoir model for the amplification of short CO_2 laser pulses.

Both parts of the model will be briefly described.

The five-temperature model is a modified version of the model described by Manes and Seguin [11]. We explicitly included intramode relaxation, so a non-Maxwellian distribution of the vibrational levels directly interacting with the laser pulse is possible. The model describes the coupling between the different vibrational modes of CO_2 and

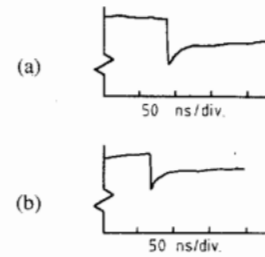


Fig. 4. Fast gain relaxation processes. Probe and pulse laser both operate on the $10.4 \mu\text{m}$ $P(20)$ transition. (a) Pulse duration up to 10 ns. (b) Pulse duration 1.0–1.5 ns.

TABLE I
SHORT RELAXATION TIME CONSTANTS (RELATED TO FERMI RESONANCE)

| SPECTRAL LINE LASER PULSE | SPECTRAL LINE PROBE LASER | RELAXATION TIME CONSTANT |
|------------------------------|------------------------------|-----------------------------|
| P(20) | P(20) | 17 ± 6 ns |
| P(20) | P(22) | 30 ± 6 ns |
| P(20) | P(24) | 26 ± 8 ns |
| P(20) | P(28) | 34 ± 7 ns |
| P(18) | P(18) | 30 ± 7 ns |
| P(18) | P(24) | 26 ± 6 ns |
| P(18) | P(28) | 25 ± 6 ns |

TABLE II
RATE CONSTANTS AT $T = 300$ K, ORIGINALLY USED IN THE NUMERICAL
MODEL ($\text{torr}^{-1}/\text{s}$), AFTER [11]

| | CO_2 | N_2 | He |
|----------|------------------|-------------------|------|
| k_{43} | 0 | $2.04 \cdot 10^4$ | 0 |
| k_{12} | $154 \cdot 10^6$ | 0 | 0 |
| k_{20} | 192 | 650 | 3270 |
| k_3 | 350 | 106 | 85 |

the vibration of N_2 by means of relaxation time constants. The vibrational energies of the modes are characterized by temperatures. T_1 , T_2 , and T_3 are the temperatures of the symmetric stretch, bending, and asymmetric stretch modes of CO_2 , respectively. T_4 is the N_2 vibrational temperature. T_5 is the rotational and translational temperature. The relaxation constants, depending on the gas composition, are given by

$$\tau_{ij} = \left\{ p \sum k_{ij,g} \cdot \Psi_g \right\}^{-1} \quad (1)$$

where p is the total pressure of the laser gas, $k_{ij,g}$ is the relaxation rate from temperature i to temperature j as a result of gas component g , and Ψ_g is the fraction of component g in the gas mixture. Table II shows values for $k_{ij,g}$ originally used in the model of [11].

Further, we use for the rotational relaxation the reservoir model, originally described by Feldman [2] and Schappert [12], which contains only one (rotational) relaxation time. We suppose that the pulse consists of M_1 P -transitions in the $00^{\circ}1$ – $10^{\circ}0$ band ($i = 1, \dots, M_1$), and $M_2 - M_1$ P -transitions in the $00^{\circ}1$ – $02^{\circ}0$ band ($i = M_1 + 1, \dots, M_2$). We then arrive at the following set of equations:

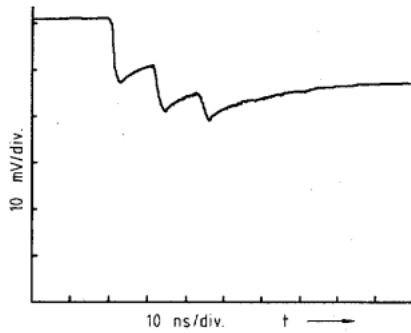


Fig. 5. Gain evolution when three nanosecond pulses are injected in the amplifier. Both lasers were operating on the 10.4 μm $P(22)$ transition.

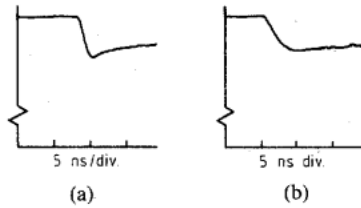


Fig. 6. Gain evolution signal showing an effect of rotational relaxation processes. (a) Pulse laser $P(20)$, probe laser $P(20)$. (b) Pulse laser $P(20)$, probe laser $P(22)$.

$$\frac{d\delta_1^J}{dt} = - \sum_{i=1}^{M_1} \delta_1^{J_i} \sigma_i \frac{I_i}{h\nu_i} \left(1 + \frac{2J_i - 1}{2J_i + 1} \right) \delta_{J,J_i} + \frac{\bar{\delta}_1^J - \delta_1^J}{\tau_R} \quad (2)$$

and

$$\frac{d\delta_2^J}{dt} = - \sum_{i=M_1+1}^{M_2} \delta_2^{J_i} \sigma_i \frac{I_i}{h\nu_i} \left(1 + \frac{2J_i - 1}{2J_i + 1} \right) \delta_{J,J_i} + \frac{\bar{\delta}_2^J - \delta_2^J}{\tau_R} \quad (3)$$

where

- J rotational quantum number of the lower laser level,
- σ_i cross section for stimulated emission of the $P(J_i)$ transition,
- I_i intensity interacting with the $P(J_i)$ transition,
- ν_i frequency corresponding to the $P(J_i)$ transition,
- δ_1^J population inversion density of a transition with rotational quantum number J in the 10.4 μm P -branch,
- δ_2^J population inversion density of a transition with rotational quantum number J in the 9.4 μm P -branch,
- δ_{J,J_i} Kronecker delta function,
- τ_R rotational relaxation time constant.

Finally, we have

$$\bar{\delta}_1^J = P(J-1)N^{00^1} - P(J) \frac{2J-1}{2J+1} N^{10^0} \quad (4)$$

$$\bar{\delta}_2^J = P(J-1)N^{00^1} - P(J) \frac{2J-1}{2J+1} N^{02^0} \quad (5)$$

where $P(J)$ is the partition fraction of the rotational sub-level with quantum number J , $\bar{\delta}_1^J$ is the equilibrium value of δ_1^J , and $\bar{\delta}_2^J$ is the equilibrium value of δ_2^J . Equations (2) and (3) are derived under the assumption that there are no ΔJ selection rules in the rotational relaxation processes.

For nanosecond pulses also, the experiments reveal that intramode relaxation may have a substantial effect on the population number densities of the lasing vibrational levels. This means that the change of these population number densities must be described by an equation that includes the intramode relaxation processes:

$$\frac{dN^{00^1}}{dt} = - \sum_{i=1}^{M_1} \delta_1^{J_i} \sigma_i \frac{I_i}{h\nu_i} - \sum_{i=M_1+1}^{M_2} \delta_2^{J_i} \sigma_i \frac{I_i}{h\nu_i} + \frac{\bar{N}^{00^1} - N^{00^1}}{\tau_{vv}^3} \quad (6)$$

$$\frac{dN^{10^0}}{dt} = \sum_{i=1}^{M_1} \delta_1^{J_i} \sigma_i \frac{I_i}{h\nu_i} + \frac{\bar{N}^{10^0} - N^{10^0}}{\tau_{vv}^1} \quad (7)$$

$$\frac{dN^{02^0}}{dt} = \sum_{i=M_1+1}^{M_2} \delta_2^{J_i} \sigma_i \frac{I_i}{h\nu_i} + \frac{\bar{N}^{02^0} - N^{02^0}}{\tau_{vv}^2} \quad (8)$$

where N^{00^1} , N^{10^0} , and N^{02^0} are the population number densities of the corresponding vibrational levels and τ_{vv}^1 , τ_{vv}^2 , and τ_{vv}^3 are the intramode vibrational relaxation time constants for the ν_1 , ν_2 , and ν_3 modes, respectively. Thus, in (2)–(8), we consider during the short period of pulse amplification only the intramode and rotational relaxation phenomena. All other relaxation processes are not important on this time scale and are neglected.

The equilibrium values \bar{N}^{00^1} , \bar{N}^{10^0} , and \bar{N}^{02^0} in (6)–(8) are calculated from the energy stored in the three vibrations. These energies correspond to temperatures. Knowing these temperatures, the equilibrium populations are easily calculated. So we need equations for the energies.

$$\frac{dE_1}{dt} = \left\{ \sum_{i=1}^{M_1} \delta_1^{J_i} \sigma_i \frac{I_i}{h\nu_i} \right\} \cdot h\nu_1 \quad (9)$$

$$\frac{dE_2}{dt} = \left\{ \sum_{i=M_1+1}^{M_2} \delta_2^{J_i} \sigma_i \frac{I_i}{h\nu_i} \right\} \cdot 2h\nu_2 \quad (10)$$

$$\frac{dE_3}{dt} = \left\{ - \sum_{i=1}^{M_1} \delta_1^{J_i} \sigma_i \frac{I_i}{h\nu_i} - \sum_{i=M_1+1}^{M_2} \delta_2^{J_i} \sigma_i \frac{I_i}{h\nu_i} \right\} \cdot h\nu_3 \quad (11)$$

where E_1, ν_1 , E_2, ν_2 , and E_3, ν_3 are the energy and the fundamental frequency of the symmetric stretch, bending, and asymmetric stretch modes, respectively.

Finally, we have

$$\frac{\partial I}{\partial t} + c \frac{\partial I}{\partial x} = c \left\{ \sum_{i=1}^{M_1} \delta_1^{J_i} \sigma_i I_i + \sum_{i=M_1+1}^{M_2} \delta_2^{J_i} \sigma_i I_i \right\} \quad (12)$$

where I is the total intensity of the pulse.

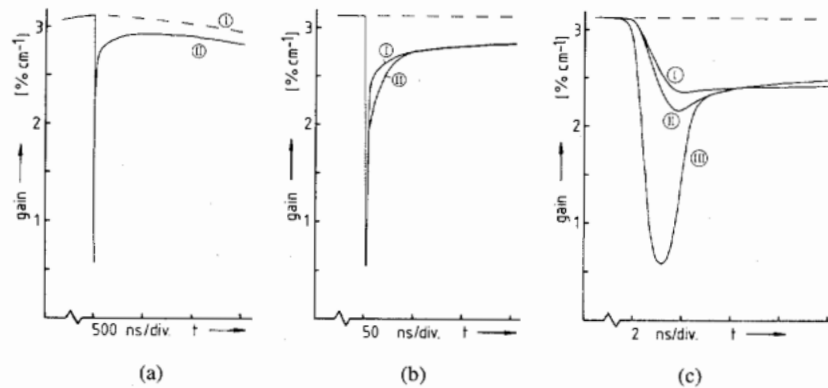


Fig. 7. Numerical results showing the evolution of the gain upon amplification of a 1 ns laser pulse on the 10.4 μm $P(20)$ transition. (a) Curve I—unperturbed gain. Curve II— $\tau_{12} = 17$ ns, $\tau_{vv} = 1$ ns. (b) Curve I— $\tau_{12} = 17$ ns, $\tau_{vv} = 1$ ns. Curve II— $\tau_{12} = 17$ ns, $\tau_{vv} = 10$ ns. (c) Curve I— $\tau_{12} = 30$ ns, $\tau_{vv} = 0.2$ ns. Curve II— $\tau_{12} = 30$ ns, $\tau_{vv} = 1$ ns. Curve III— $\tau_{12} = 30$ ns, $\tau_{vv} = 1$ ns. All curves show the $P(20)$ gain, except curves I and II in (c) where the gain of the $P(22)$ transition is shown.

IV. ANALYSIS OF THE RESULTS

The extended model can be used to simulate the amplification of a nanosecond pulse. We calculate the amplification of a laser pulse and simultaneously the evolution of the gain before and after the passage of a laser pulse through the amplifier. In the calculations of the vibrational energies and the temperatures, the Fermi relaxation constants are taken according to our measurements (see Table I), either 17 or 30 ns. The other relaxation rates that are used are given in Table II. So far, the only unknown quantities in our model are the intramode relaxation time constants. In order to obtain a good estimate of these constants, the whole process is calculated for several values, and the numerical results are then compared to the experiments. In Fig. 7, some results are plotted. We have taken the intramode relaxation time constants to be equal for the three vibrations. In this figure, the gain evolution during and after the amplification of a pulse is shown.

Fig. 7 shows that the theoretical results are similar to the experimental ones, except for the sharp dip during the amplification of the pulse. This sharp dip is due to the rotational relaxation. This phenomenon occurs on a time scale that could not be resolved by our detection system and is therefore not observed (system rise time ≈ 1 ns).

Looking at the results from Table I, we notice that in fact two values for the Fermi relaxation process are found. This is not related to rotational relaxations, as can be concluded from the results obtained with both lasers operating on the $P(18)$ transition. We cannot explain this effect. The experimental results in Fig. 4 show a fast recovery of the gain with a value that is about 35 percent of the disturbance. From the numerical results (Fig. 7(b), curve I), we notice that if intramode relaxation processes are very fast and completed shortly after amplification of a pulse, the recovery of the gain by Fermi resonance is about one-third of the remaining disturbance, in agreement with our observations (see Fig. 4). If, however, the effective

intramode relaxation time is not very fast (more than a few nanoseconds) then the recovery of the gain as a result of intramode relaxation and Fermi relaxation is about 68 percent of the remaining disturbance (see Fig. 7(b), curve II). This is not in agreement with our experimental observations.

For comparison, we notice that Stark [5] has reported a value for the rate constant of the $10^0\text{--}02^0$ process (Fermi relaxation) equal to

$$1.42 \cdot 10^5 (P_{\text{CO}_2} + 0.46 P_{\text{N}_2} + 0.054 P_{\text{H}_2})$$

(pressures in torr, $T = 400$ K).

Using his expression, we obtain for the conditions of our TEA laser a time constant of 28.5 ns. This time is quite comparable to our results (see Table I) except for the case where both lasers operate on the $P(20)$ transition.

From our experimental observations, we cannot find directly the value for the effective intramode relaxation rate. This means that this rate constant k_{vv} is larger than $6 \cdot 10^6$ ($\text{torr}^{-1}/\text{s}$). This is in agreement with the usual assumptions. Thus, intramode relaxation is very fast and may not be neglected in nanosecond pulse amplification processes.

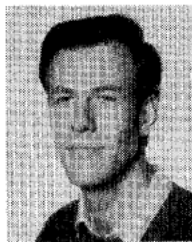
It is, however, emphasized that we did not observe intramode relaxation processes explicitly because it may be that the time constants are only a few times larger than the rotational relaxation time constants. For that reason, we calculated the decrease of the small-signal gain that could be expected in our experimental situation. For a pulse of 100 mJ input energy, it is expected that the observed gain is about 83 percent of the original value at the time both rotational and intramode relaxation are just completed. This is in agreement with the experiments, where we often observed a decrease in gain of 15 percent. This strengthens our opinion that the effective intramode relaxation time constant is less than the rise time of our detection system (≈ 1 ns).

V. CONCLUSIONS

This paper deals with an experimental and numerical investigation of the evolution of the gain upon amplification of a nanosecond laser pulse. For a multipass amplifier, it is desirable to have an optimum recovery of the gain before the pulse is amplified again. The effective intramode relaxation rate constant is estimated to be larger than $6 \cdot 10^6 \text{ torr}^{-1}/\text{s}$. The Fermi relaxation time constant in our laser was found to be $30 \pm 7 \text{ ns}$. We therefore conclude that, almost directly, (within a few nanoseconds) both rotational and intramode relaxation are complete. The energy transfer between the symmetric stretch and bending modes is completed in about 60 ns (two times the relaxation time), whereas it takes several hundred nanoseconds to transfer energy from nitrogen to the carbon dioxide asymmetrical stretch mode.

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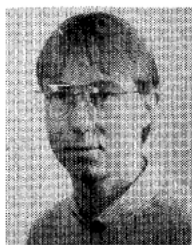
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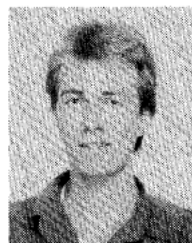
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